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Frithjof Nolting :: Head of LSC :: Paul Scherrer Institut

Time resolved X-ray experiments

PSI Master School 2017

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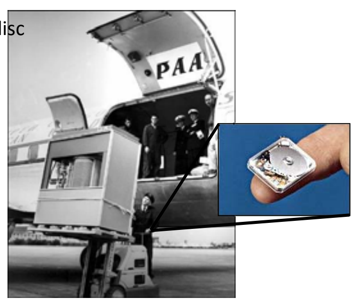
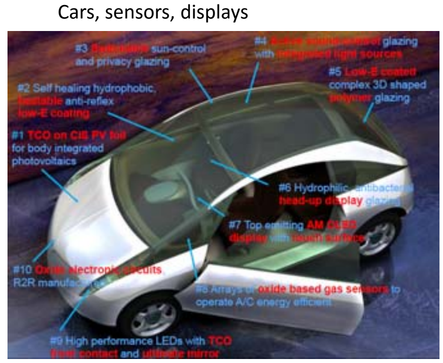
Basic research – electronic devices

Hard disc



Cars, sensors, displays



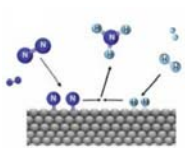
Modern communication devices are full of fascinating physics and advanced materials

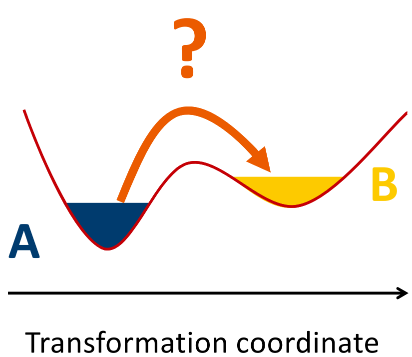
Aim of the lecture

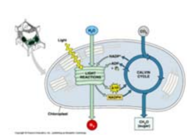
Basic concepts of time resolved measurements

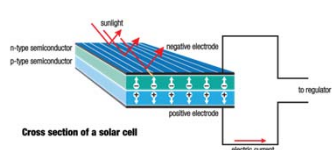
- Time scales
- How to take fast pictures
- Stroboscopic measurements/Pump probe
- Example all-optical switching
- From synchrotron to X-FEL
- Example, single shot measurement

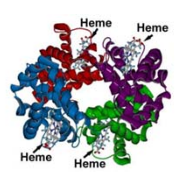
How do materials change their function?







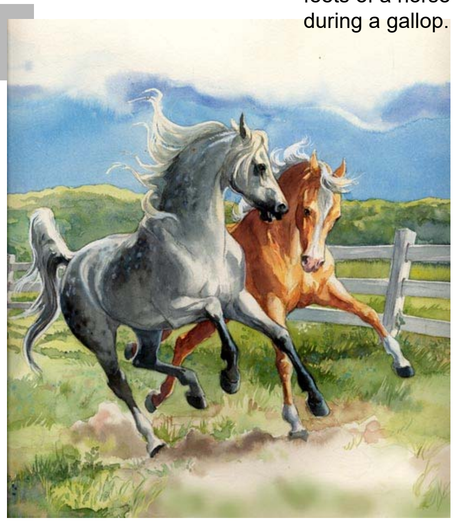




Why time-resolved?

Why time-resolved?

In 1870ths, popularly debated question: whether all four feets of a horse were off the ground at the same time while during a gallop.



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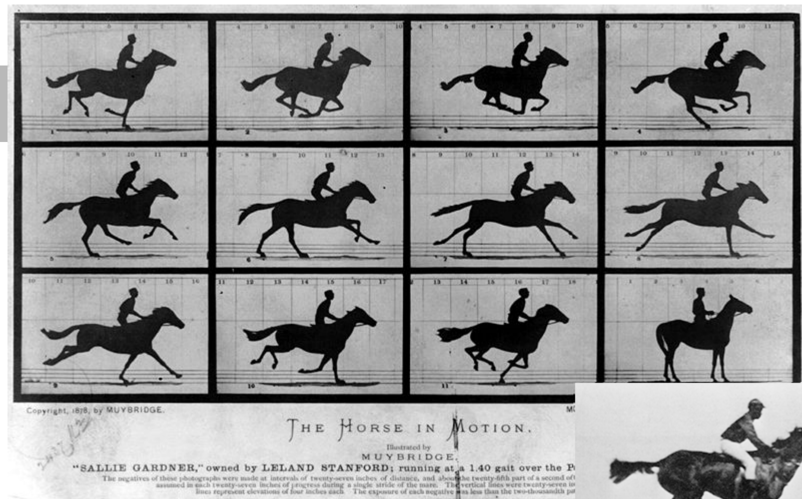
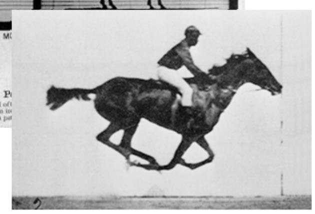
Why time-resolved?





Popularly debated question: whether all four feet of a horse were off the ground at the same time while during a gallop.

Galloping horse, animated in 2006, using photos by Eadweard Muybridge, Wikipedia

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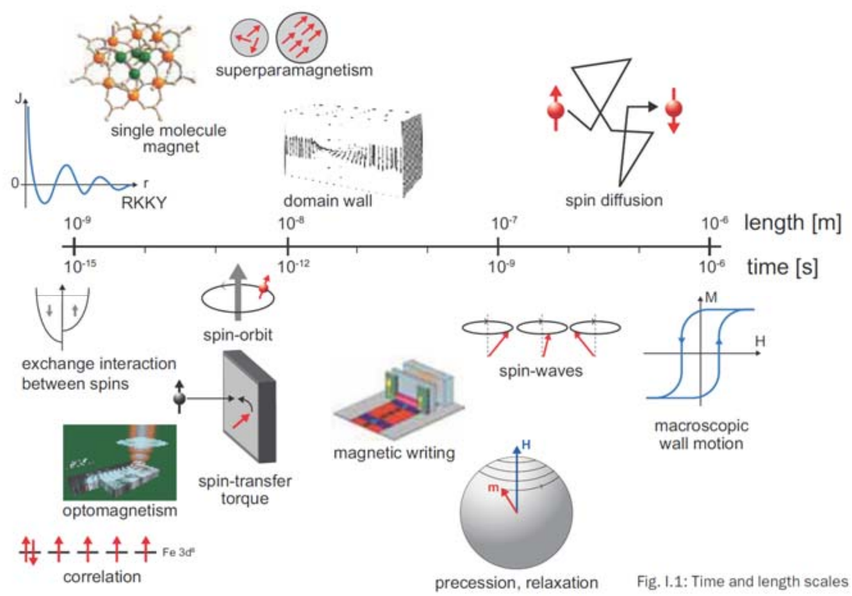
Time scales magnetism



Courtesy Bruce Patterson

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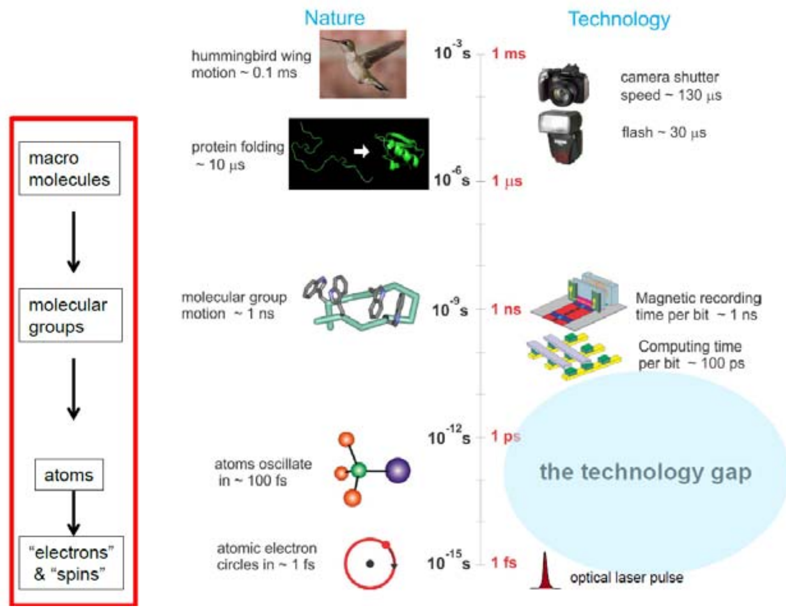
The speed of things – the smaller the faster



Courtesy Jo Stöhr

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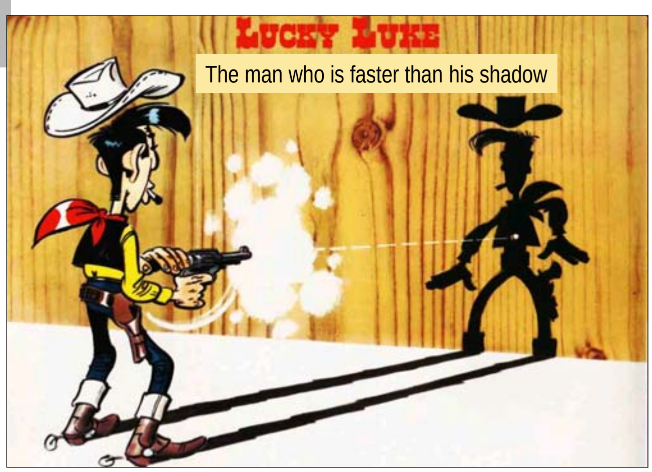
Ultrafast?

50-80 ms Blink of an eye

320 ms length of time a single frame on TV screen is shown



How much time has Lucky Luke? About 10 ns

Characteristic speeds of atoms and electrons

Atoms - speed of sounds:	1nm / 1ps
Electrons – Fermi velocity:	1nm / 1fs
Light – speed of light:	1nm / 3as

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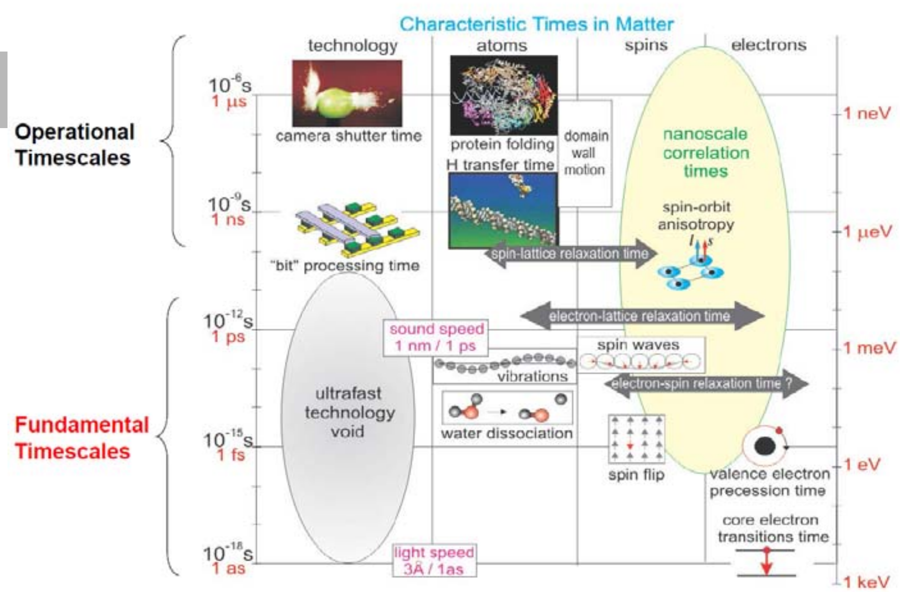
Characteristic energy and time scales in matter



Courtesy Jo Stöhr

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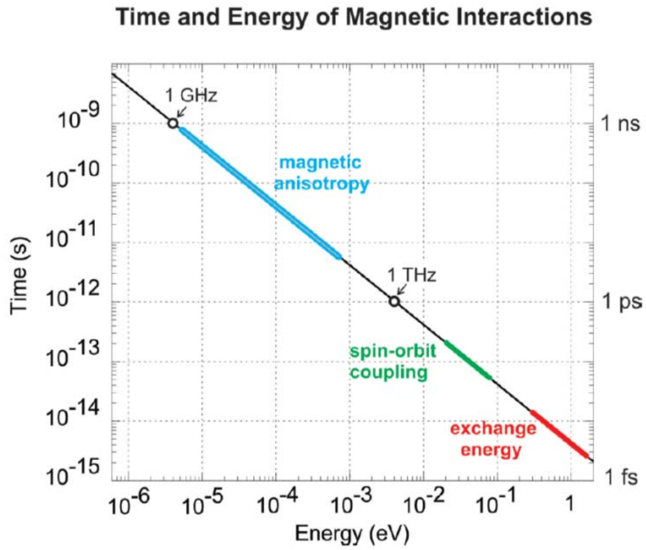
Approximate energy and time relation



Courtesy Jo Stöhr

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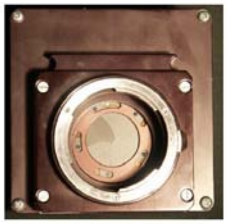
Fast photography with visible light



Fastest camera has shutter speed of 0.2 ms

- hummingbird has blurry wings
- picture typically dark because exposure is too short

Courtesy Jo Stöhr

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The trick to recording ultrafast pictures

- Use a bright flash, faster than existing shutter speed
- Capture bright reflected light flash with camera

leave shutter open, flash light is stronger than background light

light flash duration and intensity determines picture quality

Courtesy Jo Stöhr

The trick to recording ultrafast pictures

light flash duration and intensity determines picture quality

Courtesy Jo Stöhr

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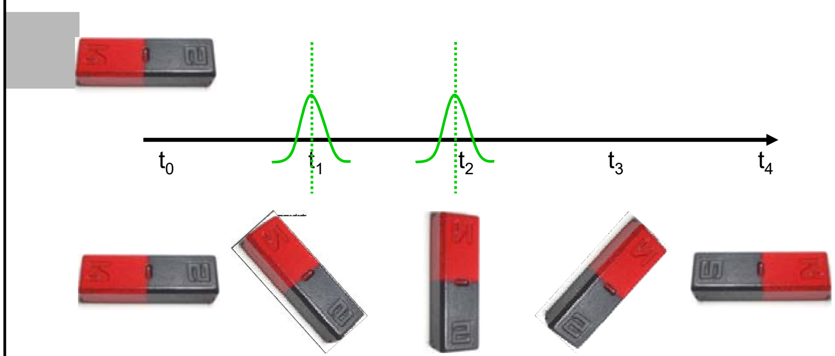
Intensity issue: stroboscopic measurement

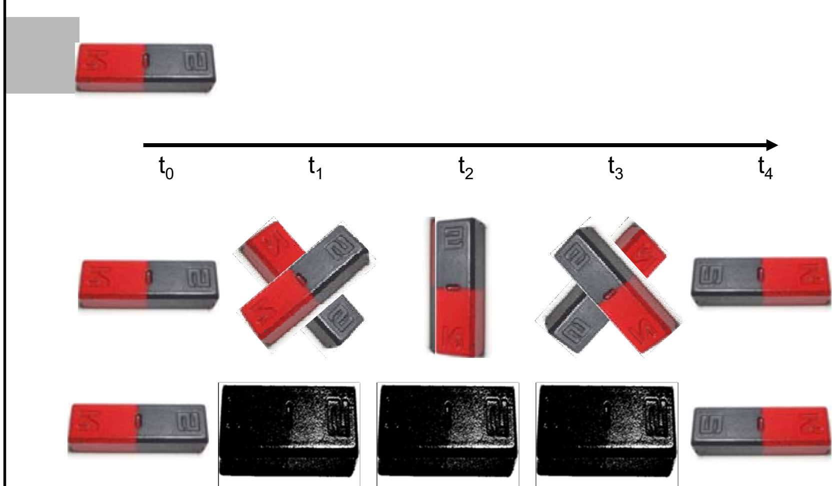


Not enough intensity for single picture

- Repeat pump-probe with fixed delay several 1000 times

Intensity issue: stroboscopic measurement

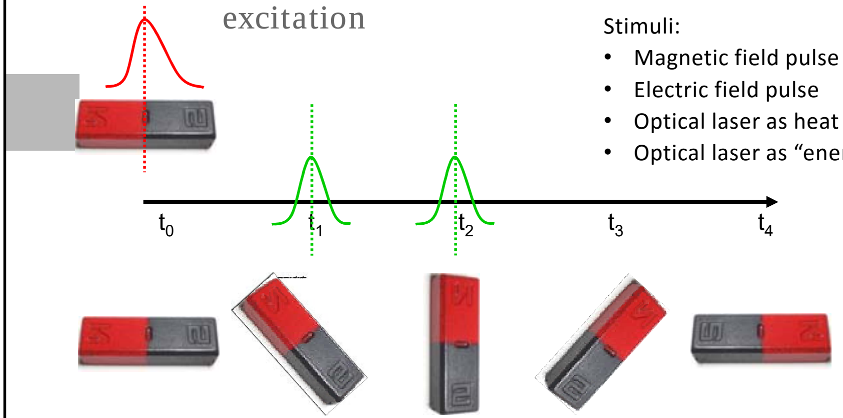
We need a repeatable effect



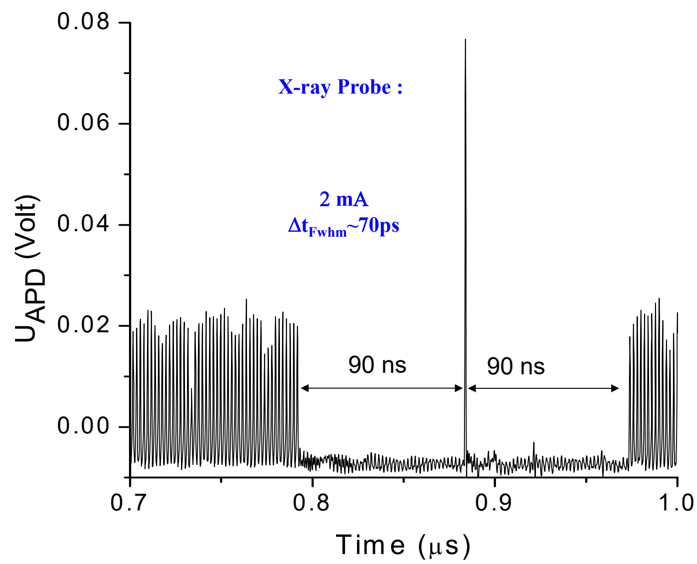
Pump – probe: study temporal response after excitation

Stimuli:

- Magnetic field pulse
- Electric field pulse
- Optical laser as heat pulse
- Optical laser as “energy” pulse



Fast dynamics: Use pulsed structure of SLS



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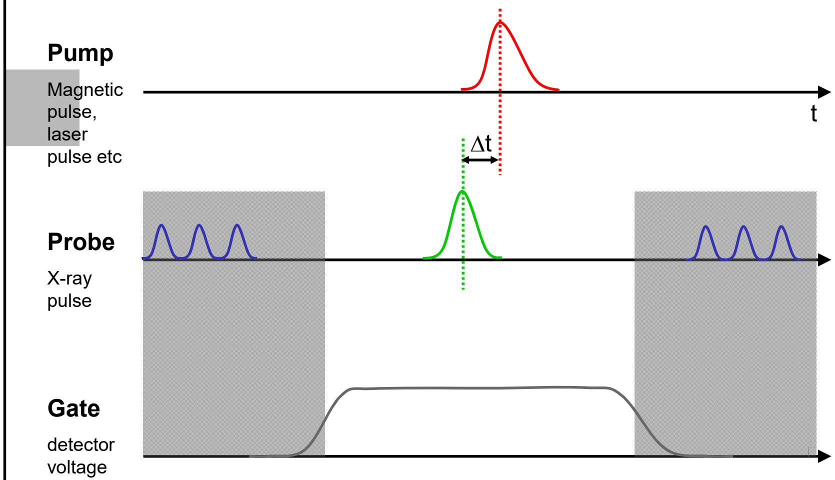
Pump - Probe



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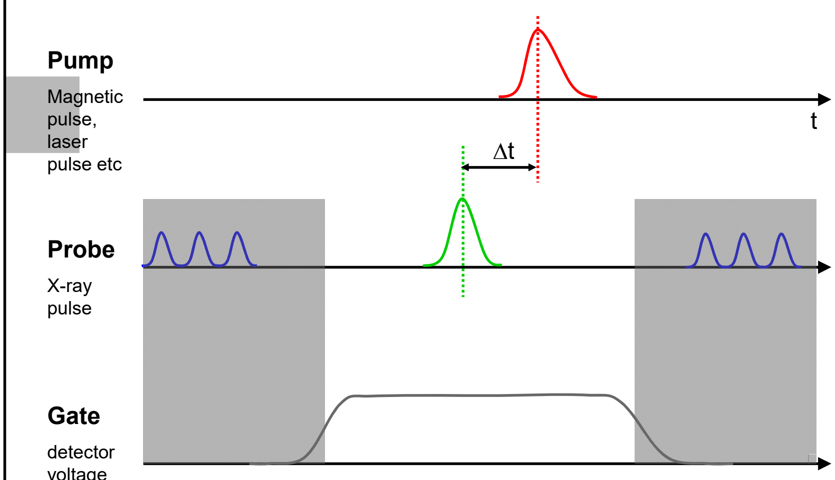
Pump - Probe



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How to change a magnetization direction

With a magnetic field

With a current

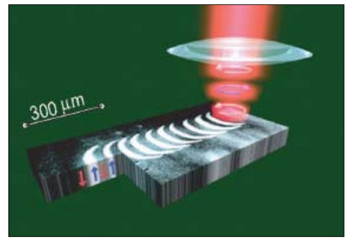
With circular polarized laser pulse

With linear polarized laser pulse
(energy pulse e.g. “heat” pulse)

Switching using a circular polarized laser pulse

GdFeCo

Stanciu et al. PRL 99, 047601 (2007)

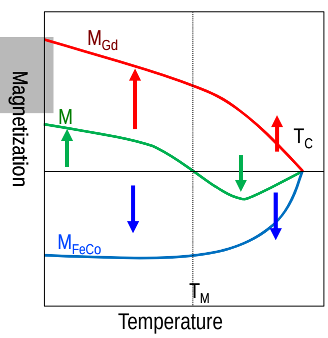


Using **circular polarized** laser pulse

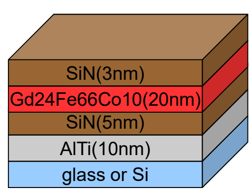
Heating up the system close to Tc

Small field for reversal needed (inverse faraday?)

The ferrimagnet: GdFeCo properties



- amorphous RE-TM alloy
- ferrimagnet
- magnetization compensation point T_M
- out-of-plane magnetic anisotropy
- strong magneto-optical properties



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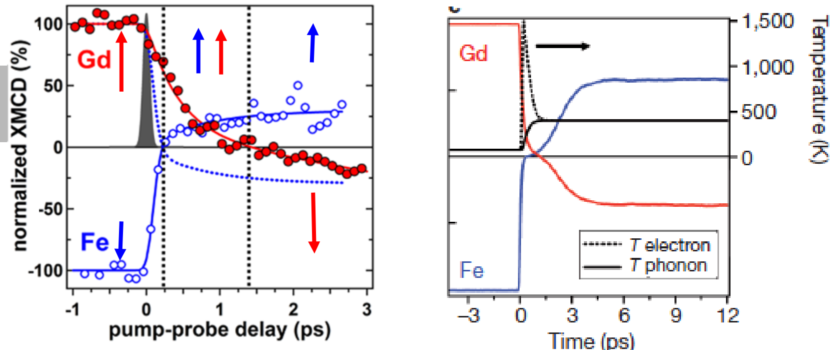
Different demagnetization time scales



I. Radu et al., Nature (2011)

- Distinct switching dynamics at the Fe and Gd sites.
- Fe switches ~ 300 fs, Gd ~ 1.5 ps. (measured in applied magnetic fields, Bessy)
- Angular momentum conservation (via exchange)

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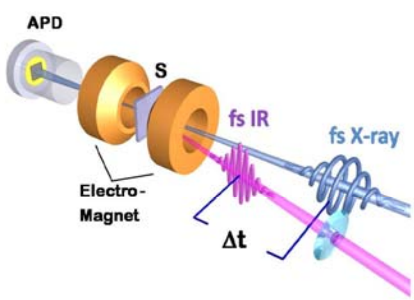
Time resolved XMCD



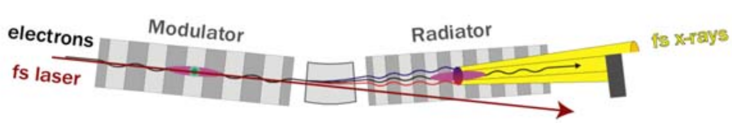
Fs-Slicing Facility @ BESSY II:

Energy range: 250-1500 eV

Variable Polarization: linear to circular

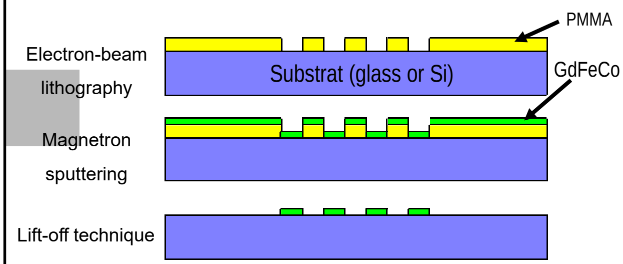
X-ray pulse lengths: 100 fs (FWHM, slicing)
10 ps (low-alpha mode)
50 ps (user mode)

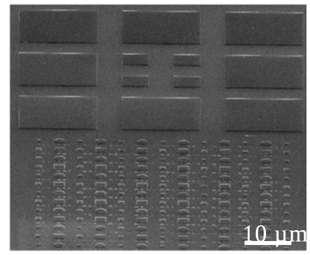
Repetition rate slicing: 6 kHz
Variable pump-wavelength UV-FIR: 220 nm – 20 μm

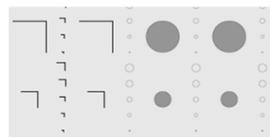


*K. Holldack et al., PRL **96**, 054801 (2006); PR ST Accel. Beams **8**, 040704 (2005)*

Nanostructuring of GdFeCo thin film







2μm down to 100 nm

Sputtering : A. Tsukamoto, A. Itoh (Nihon University, JPN)

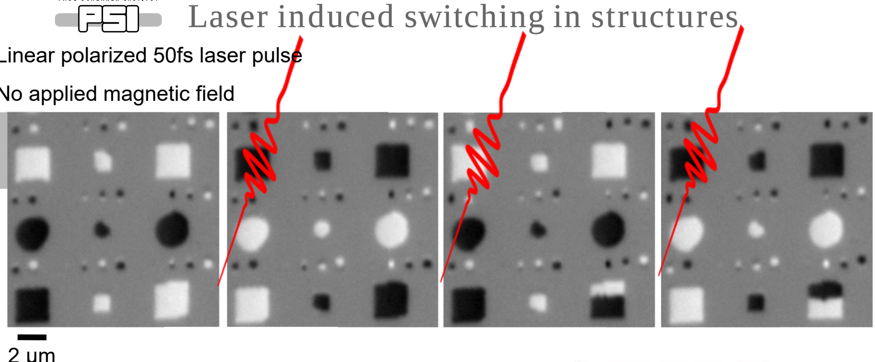
Structuring : A. Weber, E. Mengotti, L. J. Heyderman (Paul Scherrer Institut, CH)

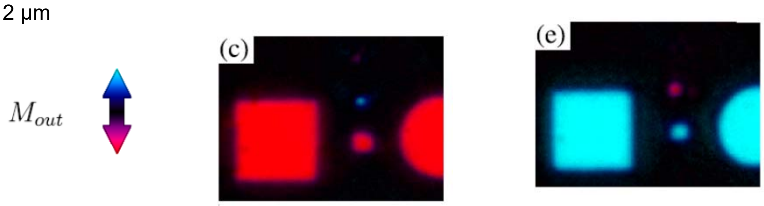
L. Le Guyader *et al.*, J. Magn. Soc. Jpn, **36**, 21 (2012)

Laser induced switching in structures

Linear polarized 50fs laser pulse

No applied magnetic field



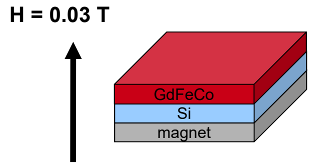


Down to 200 nm domains

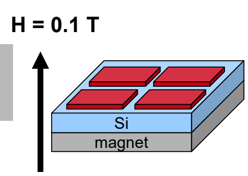
L. Le Guyader, S. El Moussaoui, M. Buzzi, R. V. Chopdekar, L. J. Heyderman, A. Tsukamoto, A. Itoh, A. Kirilyuk, Th. Rasing, A. V. Kimel, and F. Nolting, App Phys. Lett. 101, 022410 (2012).

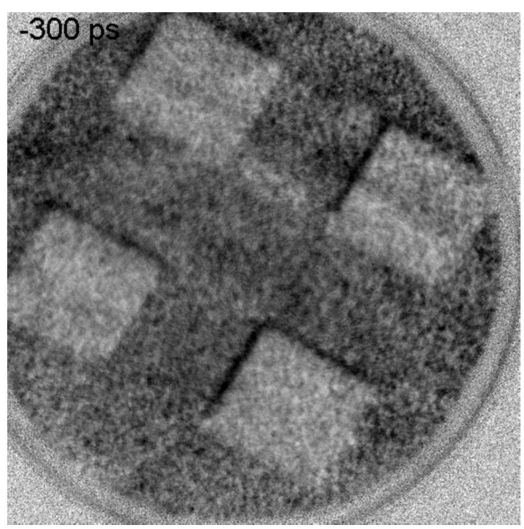
Pump – Probe?

How to reset?



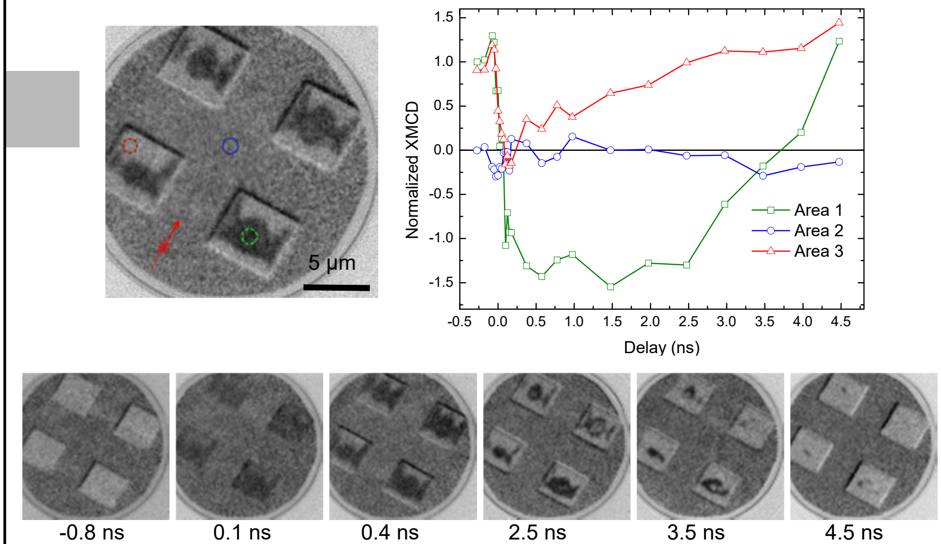
Time resolved structures



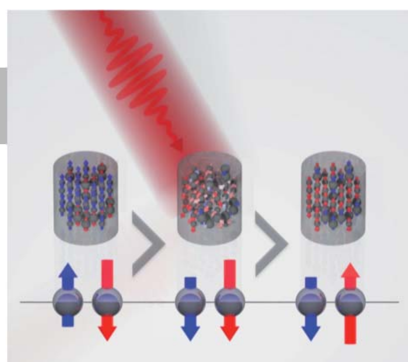


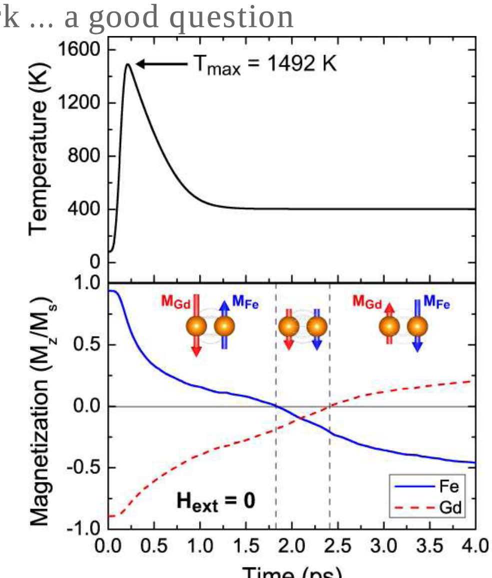
5 µm

Time resolved structures



How does it work ... a good question





Angular momentum
Different magnetization
Anti-parallel coupling
Non equilibrium

T. A. Ostler, J. Barker, R. F. L. Evans, R. Chantrell, U. Atxitia, O. Chubykalo-Fesenko, S. El Moussaoui, L. Le Guyader, E. Mengotti, L. J. Heyderman, F. Nolting, A. Tsukamoto, A. Itoh, D. Afanasiev, B. A. Ivanov, A. M. Kalashnikova, K. Vahaplar, J. Mentink, A. Kirilyuk,Th. Rasing and A. V. Kimel, Nat. Commun. 3, 666 (2012).

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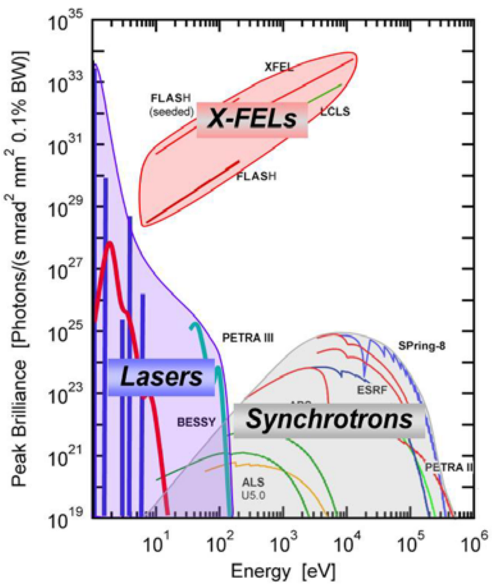
Faster?

Ultrafast flash
to overcome camera/detector speed problems

Ultrabright flash
to overcome intensity problem

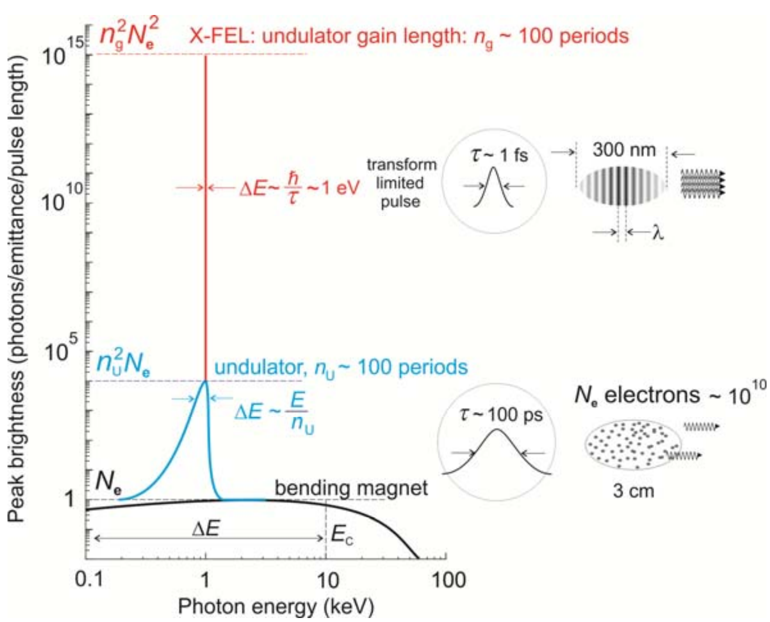
Categories of Lightsources



Synchrotron & X-FEL sources are based on electron accelerators

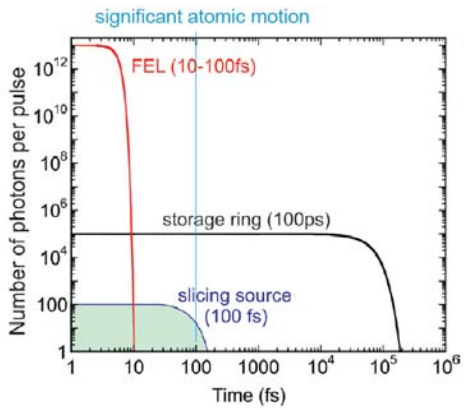
J. Ullrich, A. Rudenko, R. Moshammer
Ann. Rev. Phys. Chem. 63, 635 (2012)

X-ray source spectra



Courtesy Jo Stöhr

Pulse characteristics of X-ray sources



X-FELs offer:

Higher intensity pulses
- single pulse experiment possible

1000 x *shorter pulses*
- pulses faster than atomic motion

Coherence
- pulses are laterally coherent
- large temporally coherent fraction

But: X-FELs are fluctuating sources while storage rings are more stable

Courtesy Jo Stöhr

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Single-shot measurements

Aim:

Development of a single shot measurement technique for ultrafast processes

- Study stochastic processes
- Study processes which one cannot easy reset for stroboscopic type measurements

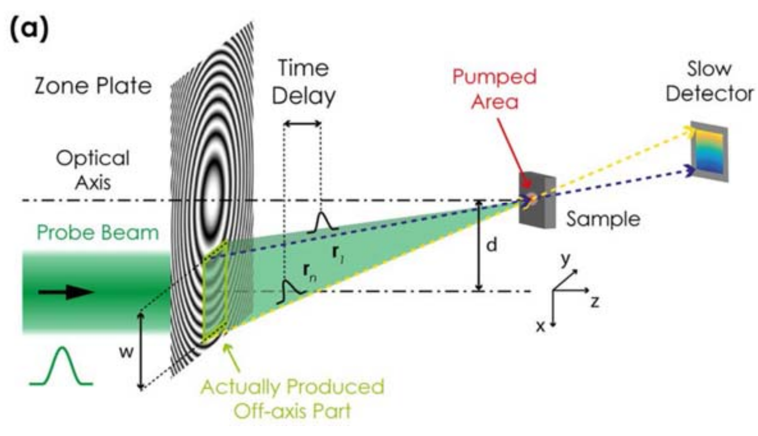
Measure early time period of ultrafast demagnetization

- Are there stochastic contributions
- Inconsistent results with different techniques

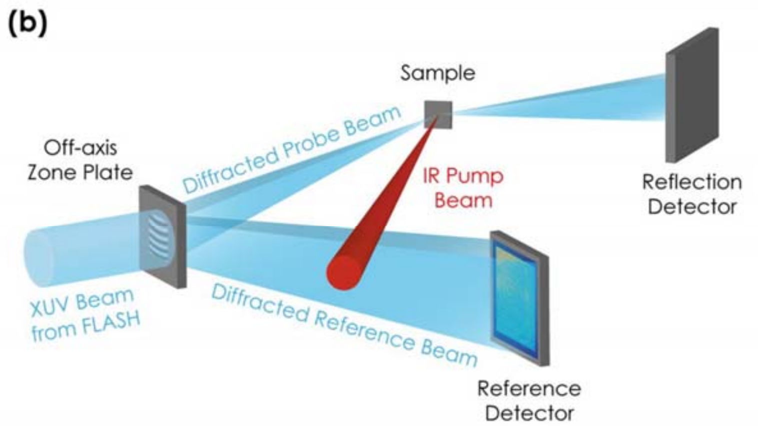
Status

- First demonstration at FLASH, Reflection and T-MOKE geometry
- Co thin film, in-plane magnetization

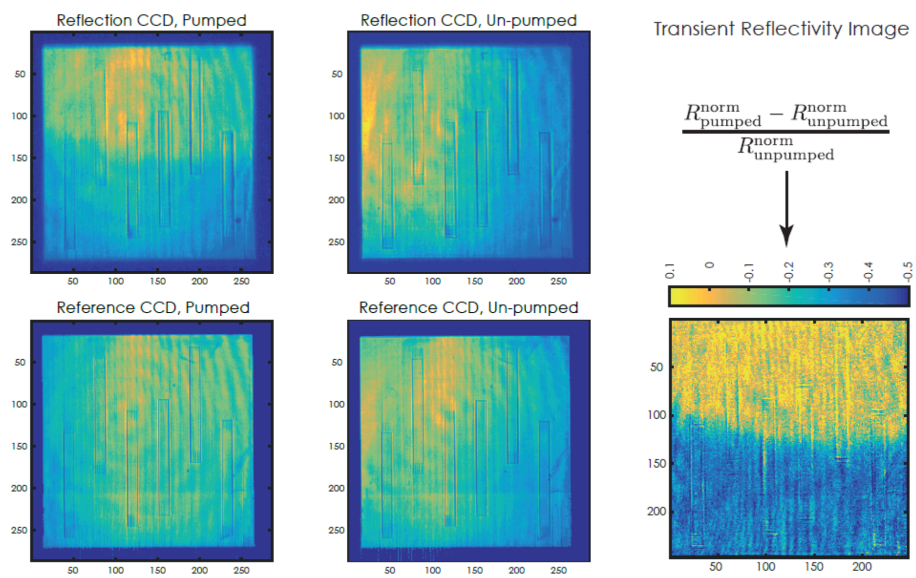
Concept: X-ray streaking



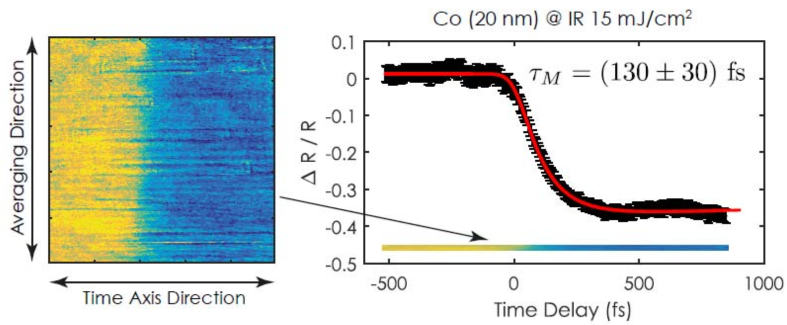
Concept: X-ray streaking



Straight forward, but …



Co thin film



$$\frac{\Delta R(t)}{R_0} = G(t) * \Theta(t)\left[A\left(1-e^{-\frac{t}{\tau_M}}\right)+B\left(1-e^{-\frac{t}{\tau_r}}\right)\right]+C$$

- Fitting parameters: τ_M, A, B, C
- Time resolution G(t) set at FWHM of 95fs.

M. Buzzi, M. Makita, L. Howald, A. Kleibert, B. Vodungbo, P. Maldonado, J. Raabe, N. Jaouen, H. Redlin, K. Tiedtke, P. M. Oppeneer, C. David, F. Nolting, and J. Lüning
Scientific Reports 7, 7253 (2017)

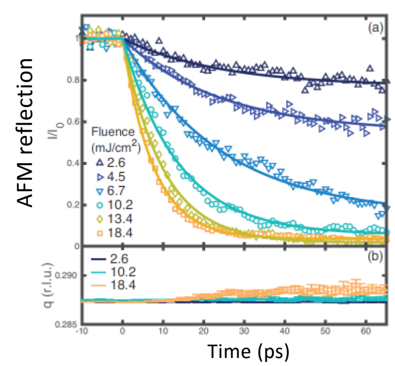
Testing ultrafast processes in condensed matter

Approach:

Pump-probe experiments to study femtosecond dynamics of

- magnetic system:
  (Resonant) x-ray diffraction (needs a FEL)
- electronic system:
  THz- or supercontinuum-probe

Example: Magnetic dynamics $TbMnO_3$



J. A. Johnson *et al.*, PRB **92**, 184429 (2015)

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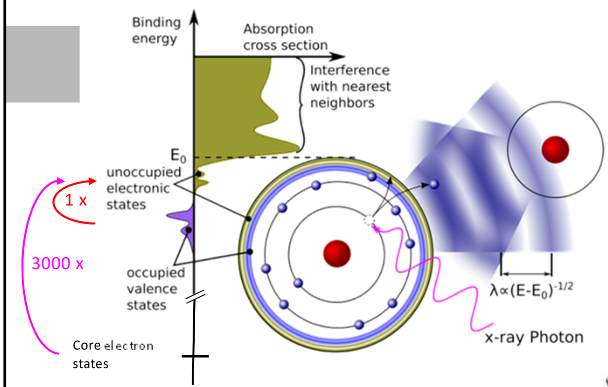
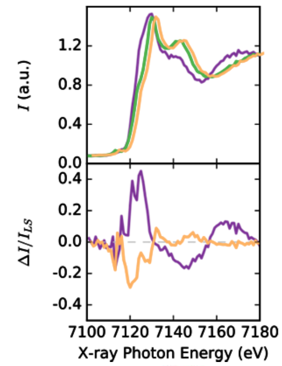
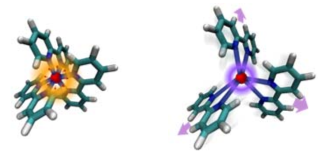
X-ray absorption spectrum → Electronic and structural information







- **Element specific**
- **Chemical potential and unoccupied states**
- **Local structure**

Charge transfer = “3+” Ox. state

High Spin = larger distance

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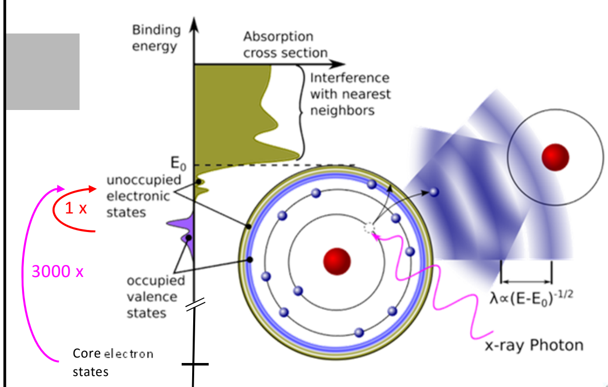
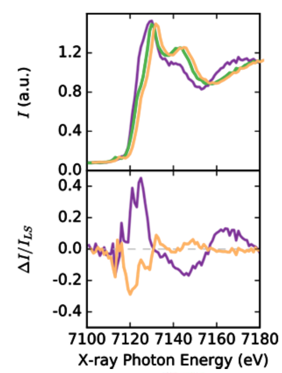
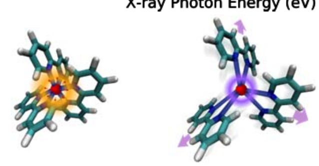
X-ray absorption spectrum → Electronic and structural information







- **Element specific**
- **Chemical potential and unoccupied states**
- **Local structure**

Charge transfer = “3+” Ox. state

High Spin = larger distance

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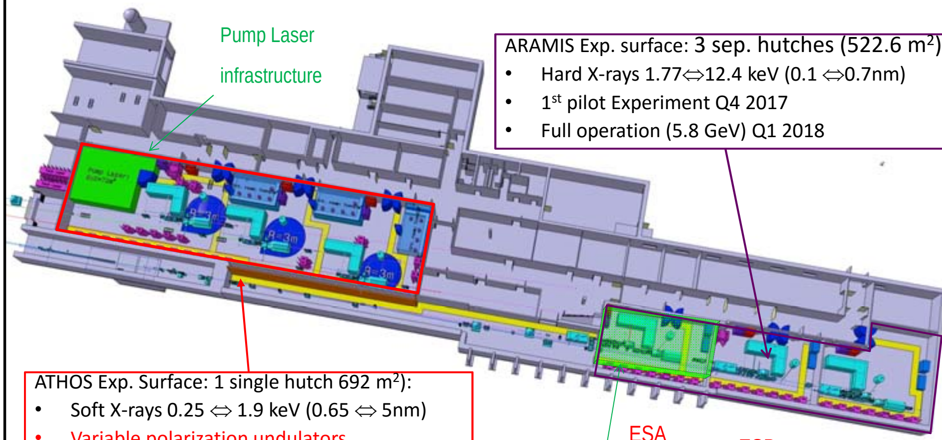
ATHOS and ARAMIS Experimental Area

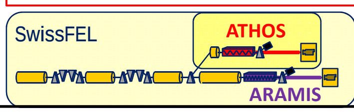


ARAMIS Exp. surface: 3 sep. hutches (522.6 m²)

- Hard X-rays 1.77⇔12.4 keV (0.1 ⇔0.7nm)
- 1st pilot Experiment Q4 2017
- Full operation (5.8 GeV) Q1 2018

ATHOS Exp. Surface: 1 single hutch 692 m²):

- Soft X-rays 0.25 ⇔ 1.9 keV (0.65 ⇔ 5nm)
- Variable polarization undulators
- Modes: SASE + self seeded + broadband + two colors + CHIC + (burst) +…
- Phase II: 2017-2020



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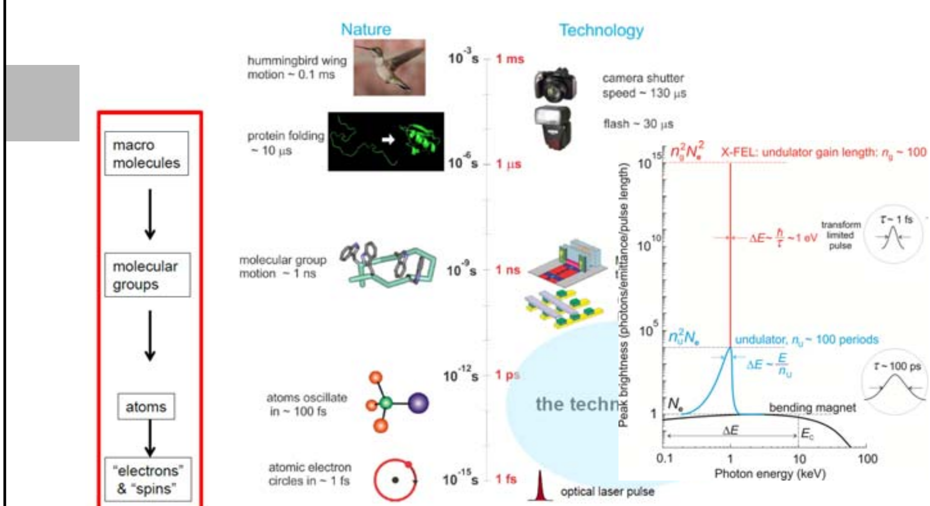
The speed of things – the smaller the faster



Courtesy Jo Stöhr